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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
10/815,879	03/31/2004	Sunil Ashitekar	135860-1	9100

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EXAMINER

BOYKIN, TERRESSA M

ART UNIT	PAPER NUMBER
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1711

DATE MAILED: 06/06/2005

Please find below and/or attached an Office communication concerning this application or proceeding.

Office Action Summary

Application No.

10/815,879

Applicant(s)

ASHTEKAR ET AL

Examiner

Terressa M. Boykin

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-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTH(S) FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If the period for reply specified above is less than thirty (30) days, a reply within the statutory minimum of thirty (30) days will be considered timely.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

Status

- 1) ☒ Responsive to communication(s) filed on 31 March 2004.
- 2a) ☐ This action is FINAL. 2b) ☒ This action is non-final.
- 3) ☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

Disposition of Claims

- 4) ☒ Claim(s) 1-28 is/are pending in the application.
- 4a) Of the above claim(s) _____ is/are withdrawn from consideration.
- 5) ☐ Claim(s) _____ is/are allowed.
- 6) ☒ Claim(s) 2, 3, 4, 11, 1, 5 -10, 14, 15, 17, 18 and 21 is/are rejected.
- 7) ☒ Claim(s) 19, 20, 22, 27 and 28 is/are objected to.
- 8) ☐ Claim(s) _____ are subject to restriction and/or election requirement.

Application Papers

- 9) ☐ The specification is objected to by the Examiner.
- 10) ☐ The drawing(s) filed on _____ is/are: a) ☐ accepted or b) ☐ objected to by the Examiner.
Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).
Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).
- 11) ☐ The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

Priority under 35 U.S.C. § 119

- 12) ☐ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).
- a) ☐ All b) ☐ Some * c) ☐ None of:
- ☐ Certified copies of the priority documents have been received.
 - ☐ Certified copies of the priority documents have been received in Application No. _____.
 - ☐ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

* See the attached detailed Office action for a list of the certified copies not received.

Attachment(s)

- ☒ Notice of References Cited (PTO-892)
- ☐ Notice of Draftsperson's Patent Drawing Review (PTO-948)
- ☒ Information Disclosure Statement(s) (PTO-1449 or PTO/SB/08)
Paper No(s)/Mail Date 2-23-05.
- ☐ Interview Summary (PTO-413)
Paper No(s)/Mail Date. _____.
- ☐ Notice of Informal Patent Application (PTO-152)
- ☐ Other: _____.

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Claim Rejections - 35 USC § 102

The following is a quotation of the appropriate paragraphs of 35 U.S.C. 102 that form the basis for the rejections under this section made in this Office action:

A person shall be entitled to a patent unless –

(b) the invention was patented or described in a printed publication in this or a foreign country or in public use or on sale in this country, more than one year prior to the date of application for patent in the United States.

Claims 1, 5, 6, 7, 8, 9, 10, 14, 15, 17, 18, 21, 23-26 are rejected under 35 U.S.C. 102(b) as being anticipated by USP 4644086, note col. 1 lines 57 – 58, cols. 2-4, examples 1 - 9 and claims.

Applicants' claims are directed to a continuous process comprising contacting a mixture comprising a dihydroxy aromatic compound, water and an alkylating agent with a catalyst system in the presence of a flowing carrier gas.

USP 4644086 discloses an improved gaseous phase catalytic process for the orthomethylation of phenol compounds having at least one hydrogen atom in the ortho position which employ oxide metal mixtures. These oxide mixture catalysts exhibit high activity and selectivity for ortho-alkylation over prolonged time periods. For example, the reference discloses production of high yields of orthocresol from phenol, 2,6-dimethylphenol from phenol and/or orthocresol 2,3,6-trimethylphenol from metacresol, and 2,3,5,6-tetramethylphenol from 3,5-dimethylphenol. Although, it is noted that that the reference refers to phenols in general, such term is not considered limiting and may encompass di, tri, phenols. For example, in the USP 4644086 reference's specification in col. 1 lines 57 – 58 it states that the phenol compounds *have at least one hydrogen*

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atom in the ortho position which would include additional moieties, i.e. at least one hydrogen atoms in the meta or para position, consequently anticipating applicants dihydroxy compounds as well.

The catalysts useable in the process of this invention are obtained by combining (a) iron oxide, with (b) molybdenum oxide and/or tungsten oxide in conjunction with (c) one or more of the oxides of the alkaline earth metals, specifically magnesium, calcium or barium, or of the rare earths specifically lanthanum or cerium, or of manganese. Combinations of the various alkaline earths, combinations of the various rare earths, combinations of the alkaline earths with the rare earths all optionally in combination with and/or manganese are also contemplated in the invention.

It is also disclosed that the catalysts system undergoes calcination. For example, the catalysts of reference are prepared according to conventional methods, for example by intimate mixing of the oxide components having a particle size range of preferably about 1 to 10 μm (micrometers) or preferably by coprecipitation of the hydroxides and oxides, respectively, from aqueous solutions of the metallic salts.

Coprecipitation can be accomplished with, for example, molybdenum in the form of ammonium molybdate, or tungsten in the form of ammonium tungstate, coprecipitated with bases, such as aqueous ammonia solution, alkaline solutions or *alkali carbonate solutions*. Note applicants claim 10. The iron hydroxide, for example iron (III) hydroxide precipitate, containing the oxides

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and/or hydroxides of the additive components in uniform distribution, is washed with water to remove foreign ions and excess base that may be present, dried at about 130.degree. C., and ground into a powder. The pulverulent oxide mixture, to which may optionally be added up to 10% by weight of an inert additive, such as graphite, can be pressed or shaped into molded elements of a suitable size, for example into tablets or extruded shapes. After *calcining* at temperatures of about 400.-500 C., the molded catalyst articles are ready for the gaseous phase reaction of the phenols with methanol. In a preferred embodiment, the molded elements are filled into reaction tubes having internal diameters of up to 50 mm, preferably up to 32 mm, said tubes being immersible in a thermostatically controlled heating bath. The reactants (phenolic compound and methanol) are vaporized in a specific molar ratio prior to entering the catalyst-filled reaction tube; suitably by feeding a solution of the phenolic compound in the stoichiometric amount of methanol to a vaporizer/preheater and conducting the vapor mixture existing therefrom over the catalyst.

With regard to the pore former, applicants state in the specification page 5 lines 1-2 that the pore former used in the catalyst system is *a substance capable* of aiding the formation of pores in the catalysts. As noted above, an inert additive, such as graphite, may be added to the oxide mixture which is then pressed or shaped into molded elements of a suitable size. The physical addition of the graphite additive would cause pores in the catalyst system and thus would render the claimed invention anticipated.

As the alkylating agent the reference employs methanol or methyl alcohol.

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With regard to applicants claim 15 note that the reference discloses that the phenols are reacted with methanol in a molar ratio of about 1:1 to 1:10 which overlaps applicants claimed invention.

With regard to applicants' Claims 14, 15, 17 and 21 as well as claims , the reference notes that the process and catalyst for the orthomethylation of phenols having at least one free ortho position, wherein the phenols are reacted with methanol in a molar ratio of about 1:1 to 1:10 at a *temperature of about 250 - 340 C.* in the gaseous phase under a pressure of about 1-4 bar absolute and *with rates per unit volume (LHSV) of about 0.05-3 h.⁻¹* in the presence of an iron oxide catalyst which overlaps applicants claimed weighted hourly space velocity of 0.1 to 10 as well as the molar ratios and temperature range used therein.

As noted in previous office actions, applicants' discussion correctly states that phenols are *generally* considered to be monohydroxylated and may be further substituted with alkyl groups as known in the art. However, generally considered does not exclude the phenols from being including dihydroxy aromatic compounds or substituted compounds. Applicants claimed invention does not exclude the phenols of the prior art.

Additionally, the recited "optionally be added up to 10% by weight of an inert additive, such as graphite, can be pressed or shaped into molded elements of a suitable size, for example into tablets or extruded shapes" may well be in the form of pores. Applicants' definition or limitation of the meaning of pore former

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has not been explicitly stated. As written, a "pore former" would include any additive which formed or resulted in occlusions or holes in the composition.

Thus, the reference discloses a continuous process comprising the same components and steps as claimed by applicants. There appears to be no significant difference between the reference and that which is claimed by applicant(s). Any differences not specifically mentioned appear to be conventional. Consequently, the claimed invention cannot be deemed as novel and accordingly is unpatentable.

35 USC 103

The following is a quotation of 35 U.S.C. 103(a) which forms the basis for all obviousness rejections set forth in this Office action:

(a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negated by the manner in which the invention was made.

Claims 2, 3, 4, 11, are rejected under 35 U.S.C. 103(a) as being unpatentable over USP 4644086 see abstract, col. 1 lines 55-59, col. 2 lines 22 through 59, examples 1-9, and claims 1-6 and 12.

With regard to claims 2, 3 and 4 the reference discloses a prepared from the same components as claimed by applicants except for the particular dihydroxy aromatic compound such as hydroquinone etc. however, as noted above the term phenol in the particular reference encompasses all types of phenols including di and tri phenols. Note col. 1 lines 55-59. Thus, it would have been obvious to one having ordinary skill in the art at the time the invention was made

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to employ the specific dihydroxy aromatic compounds as claimed since such are included under phenols and the specification has not given any unexpected findings resulting therefrom.

With regard to claim 11, the reference discloses the claimed invention prepared from the same components as claimed by applicants except for the particular copper moiety employed therein. Note, however in col. 2 lines 22 through 59 demonstrates that metal oxides are used in the invention. The reference notes preferred metals which are found in lines 53-68. Nevertheless, copper is naturally occurring as a metal oxide and would thus be included therein. Thus, it would have been obvious to one having ordinary skill in the art at the time the invention was made to employ as acatalyst, copper since such is infact a naturally occurring metal oxide and would thus meet the requirements therein.

Consequently, the claimed invention cannot be deemed as unbvious and accordingly is unpatentable.

Claims 12 and 13 are rejected under 35 U.S.C. 103(a) as being unpatentable over US 20050004407 and US 20030194366 respectively.

With regard to claim 12, the reference discloses the claimed invention prepared from the same components as claimed by applicants except for the use of waxes and polysccharide as the pore former, however, note that **US 20050004407** discloses catalyst methods for the alkylation of hydroxyaromatic compounds. It states that the metal oxide precursor mixture further comprises a pore former. The pore former may be described as a substance capable of aiding

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the formation of pores in the catalyst. The pore former may comprise a wax, polysaccharide, or a combination of the foregoing. Useful waxes include paraffin wax, polyethylene wax, microcrystalline wax, montan wax, and the like, as well as combinations comprising at least one of the foregoing waxes. See claim 9.

With regard to claims 13 note that **US 20030194366** discusses that polyethylene glycol may be used as a pore former. It states that the pore size and pore-size distribution of a given catalyst can be increased by the addition of pore-forming precursor materials to the metal oxide powders, such as hydroxymethylcellulose or polyethylene glycol, which will burn away during calcination, leaving behind larger pores.

Thus, it would have been obvious to one having ordinary skill in the art at the time the invention was made to employ as the pore formers each of the moieties above in order to aid in the increase of the catalytic activity, increased pore size and would thus meet the requirements therein.

Claim Rejections - 35 USC § 102

The following is a quotation of the appropriate paragraphs of 35 U.S.C. 102 that form the basis for the rejections under this section made in this Office action:

A person shall be entitled to a patent unless –

(b) the invention was patented or described in a printed publication in this or a foreign country or in public use or on sale in this country, more than one year prior to the date of application for patent in the United States.

Claims 1, 5, 6, 7, 8, 9, 10, 14, 15, 16, 17, 18, 21, 23-26 are rejected under 35 U.S.C. 102(b) as being anticipated by USP 4661638, note col. 1 lines 20-65, cols. 3 lines 9-14, 41-53 and col. 4 lines 23-62 and claims.

US 4661638 a catalyst derived from mixture of manganese hydroxide and magnesium-containing material, and method of use in ortho-alkylation of phenols

A catalyst precursor prepared by precipitating manganese hydroxide from caustic solution and then mixing the precipitate with a magnesium-containing material is described. The catalyst precursor can be calcined to an active form, and the resulting catalyst can be used to effect or facilitate the ortho-alkylation of phenolic compounds in vapor phase reactions. Superiority of performance is demonstrated with respect to a catalyst derived from a precursor in which manganese hydroxide is precipitated (sometimes referred to as "co-precipitation") in the presence of a magnesium compound, rather than formed separately.

After being shaped, the catalyst precursor is activated for use by being subjected to a calcining treatment under time and temperature conditions sufficient to produce an active catalyst. Typically, the treatment involves heating the precursor to a temperature of at least 300.degree. C., or sufficient to convert the magnesium and manganese compounds to a mixture of oxides. Temperatures of between approximately 350 and 500.degree. C. for a period of about 34 hours are preferred, but temperatures as high as 550.degree. C. may be used. Calcining can be effected in a variety of environments, including air, an inert gas, e.g., nitrogen, or under vacuum. The calcination treatment may be carried out prior to loading into a reactor, or alternatively, in situ in the reactor itself, and optionally in the presence

of a feed stream of the reactants.

Phenolic compounds containing alkyl substituents in the ortho position on the ring are useful as starting materials for the preparation of polyphenylene oxide resins. In general, these compounds are prepared by catalyzed processes in which one or more phenolic compounds are reacted with an alkyl alcohol in the vapor phase.

By way of illustrating the practice of the process, any one or a mixture of phenols having an ortho hydrogen is vaporized and, together with an alkyl alcohol, is passed through a reactor heated to a temperature of at least 300.degree. C., preferably from about 400 .degree. to 500.degree. C., and containing a catalyst prepared as described. For the most favorable results, it is advisable to use at least one mole of the alkyl alcohol, and preferably from one to three moles, for each ortho position on the phenol to be alkylated. For example, if phenol, which has two ortho hydrogens per molecule, is to be methylated to produce 2, 6-xlenol in optimum yields, it is desirable to employ from two to six moles of methanol for each mole of phenol, the larger yields being obtained with use of the higher ratios of methanol to phenol.

Objected Claims

Claims 19, 20, 22, are objected to as being dependent upon a rejected base claim, but would be allowable if rewritten in independent form.

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Correspondence

Please note that the cited U.S. patents and patent application publications are available for download via the Office's PAIR. As an alternate source, all U.S. patents and patent application publications are available on the USPTO web site (www.uspto.gov), from the Office of Public Records and from commercial sources. Applicants may be referred to the Electronic Business Center (EBC) at <http://www.uspto.gov/ebc/index.html> or 1-866-217-9197.

Any inquiry concerning this communication or earlier communications from the examiner should be directed to Examiner Terressa Boykin whose telephone number is 571 272-1069. The examiner can normally be reached on Monday through Friday from 6:30am to 3:00pm.

The fax phone number for the organization where this application or proceeding is assigned is 703-872-9306. The general information number for listings of personnel is (571-272-1700). Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see <http://pair-direct.uspto.gov>. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free).

tmb



Examiner Terressa Boykin

Primary Examiner

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